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ORIGINAL PAPER

Residential Proximity to Metal-Emitting Industries and Toenail Metal Concentration in the US Gulf States

Joyce J. Y. Lin[1] · **Emily J. Werder**[2] · **Kaitlyn G. Lawrence**[2] · **W. Braxton Jackson II**[3] · **Dale P. Sandler**[2] · **Aisha S. Dickerson**[4] · **Lawrence S. Engel**[2,5] · **Ana M. Rule**[1]

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Abstract

The US Gulf region is heavily reliant on metal-emitting petrochemical and manufacturing industries. We sought to characterize associations between residential proximity to metal-emitting sites and toenail metal concentration in men from four US states along the Gulf of Mexico with particular attention to potential differential exposure burden by race. We measured toenail concentrations of arsenic, chromium, lead, manganese, mercury, and selenium using inductively coupled plasma mass spectrometry in 413 non-smoking men from the Gulf Long-term Follow-Up Study (2011–2013). Point sources of industrial metal emissions were identified using the US EPA's National Emissions Inventory (NEI) database and mapped to geocoded participant residential addresses. For each metal, we examined relationships between toenail metal concentrations and linear distance to the nearest metal emitting site, inverse distance weighted number of emissions sites, and inverse distance weighted volume of air metal emissions within 30 km radial buffers of participant residences using multivariable linear regression. Results were stratified by self-reported race. Compared to self-identified White participants, Black participants lived closer to NEI sites but had 23–70% lower toenail concentrations of arsenic, chromium, mercury, manganese, and selenium adjusting for personal/behavioral factors. Toenail lead concentration was positively associated with residential proximity to lead-emitting NEI sites though the relationship was significantly attenuated after adjustment for neighborhood-level socioeconomic factors such as poverty level and age of housing stock. Residential proximity to lead-emitting NEI sites in the Gulf region is associated with a higher body burden of lead as measured in the toenail. This relationship may be driven in part by non-NEI factors related to residence in industry-adjacent neighborhoods. Further research into dietary/occupational exposures is needed to explain the unexpected racial disparities in metal body burden in this population.

Keywords Metals · Exposure assessment · Environmental justice · Toenail biomarkers

✉ Joyce J. Y. Lin
Jlin103@jhu.edu

[1] Department of Environmental Health and Engineering, Johns Hopkins Bloomberg School of Public Health, 615 N. Wolfe Street, Baltimore, MD, USA

[2] Epidemiology Branch, National Institute of Environmental Health Sciences, National Institutes of Health, Research Triangle Park, NC, USA

[3] Social & Scientific Systems, Inc., a DLH Holdings Corp., Durham, NC, USA

[4] Department of Epidemiology, Johns Hopkins Bloomberg School of Public Health, Baltimore, MD, USA

[5] Department of Epidemiology, University of North Carolina Gillings School of Global Public Health, Chapel Hill, NC, USA

Introduction

Pollutant metals and metalloids, hereafter referred to as "metals", exist ubiquitously in the environment but concentrate in densely populated areas as a result of anthropogenic emissions from industry, agriculture, fossil fuel combustion, and waste disposal (Tchounwou et al. 2012). Since these elements do not degrade, their accumulation in the environment greatly increases the risk of chronic human exposure. The degree of metal toxicity is determined by the chemical type, dose, and route of exposure, but a wealth of evidence points to numerous adverse health effects associated with a wide range of metal exposures across the life course (Jusko et al. 2008; Attreed et al. 2017; Vahter et al. 2002; Wright and Baccarelli 2007). Since metal exposures often have anthropogenic origins, the rapidly evolving commercial uses for metals in industrial processes have raised concerns about chronic metal exposure for communities residing proximal to industrial areas (Tchounwou et al. 2012).

The concentration of industrial operations in the US Gulf of Mexico region presents a heightened risk of chronic ambient metal exposure. Metal byproducts from large-scale chemical, plastics, paper, and electrical manufacturing constitute some of the largest point sources of ambient environmental metal exposure in the Gulf region (Hassaan et al. 2016; Ha et al. 2014). In 2014, across Louisiana, Alabama, Mississippi, and Florida, more than 23 types of industries released more than 129 million pounds of various metals directly into the air, soil, and water (US EPA 2013). This concentration of industries, coupled with the Gulf region's extensive history of racial segregation, raises concerns of disproportionate metal exposures which may further exacerbate existing health disparities in the region. While historically marginalized and low-income communities have been shown to bear disproportionate burdens of environmental pollution in the US (Apelberg et al. 2005; Hajat et al. 2015; Trottier et al. 2023; Jones et al. 2022), few studies have assessed the impact of industrial emissions on human metal exposure using biomonitoring. To our knowledge, no such studies have been conducted in the uniquely vulnerable US Gulf region.

In this study, we examined the relationship between residential proximity to industry-reported air metal emissions and toenail metal concentration in a multi-state sample of men from the Gulf Long-term Follow-up (GuLF) Study. Analyses were stratified by self-reported race in consideration of potential metal exposure disparities related to the persistent effects of the area's extensive history of racial segregation.

Methods

Study Population

The Gulf Long-term Follow-up (GuLF) Study (2011–2013) is a large prospective cohort study ($n = 33{,}608$) of short- and long-term health effects related to oil spill exposures from the 2010 *Deepwater Horizon* (DWH) disaster (Kwok et al. 2017). Participants comprise individuals who either worked on the oil spill for at least one day (oil spill cleanup workers) or who took part in mandatory worker safety training but did not work on the spill (non-workers). Details about GuLF Study enrollment and cohort follow-up have been previously published (Kwok et al. 2017; Engel et al. 2017).

This research was conducted in a sample of 413 non-smoking men from the larger GuLF Study who provided toenail samples at a clinical exam visit 2 – 6 years (median 4.6 years) after the end of reported cleanup from the *DWH* disaster. Current smokers at the time of toenail sample collection were excluded to maximize the sensitivity of toenail samples to the ambient industrial metal exposures of interest. Detailed selection criteria for the subsample in this study have been reported previously and participants comprise a subset of GuLF participants with other collected biomarkers to maximize information overlap (Lin et al. 2023). Briefly, between August 2014 and June 2016, 3,401 individuals who lived within 60 miles of study clinics in Mobile, Alabama, or New Orleans, Louisiana participated in a clinical exam in which trained examiners collected health, diet, work history, and residential address, as well as toenail biospecimens, anthropometric measures, and neurobehavioral test results. The geographic distribution of participants in the analytic sample is shown in Fig. 1. Toenail samples were self-collected by study participants using stainless steel clippers, placed in paper envelopes, and stored at room temperature in the GuLF Study biorepository until analysis in 2021. Of participants who completed a neurobehavioral exam and provided sufficient toenail samples ($n = 2{,}734$), we included those with previously measured liver and kidney function/injury biomarkers, selected on the basis of oil spill exposures ($n = 679$), to maximize GuLF Study biomarker overlap. We further excluded self-reported current smokers to focus our analysis. This resulted in a final analytical population of 413 participants in this study.

Toenail Metals Analysis

Toenail samples were analyzed for 18 metals/metalloids (aluminum (Al), antimony (Sb), arsenic (As), cadmium (Cd), calcium (Ca), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), magnesium (Mg), manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), selenium (Se), vanadium (V), and zinc (Zn)) using inductively coupled plasma mass spectrometry (ICP-MS). Roughly 1 – 2 toenail clippings (median 25 mg) were randomly selected from each participant's total sample for metals analysis. This sub-sampling method maximizes sample conservation and has demonstrated reliable results in previous findings (Lin et al. 2023).

Details about the toenail metal analysis process are described elsewhere (Lin et al. 2023). In brief, toenail samples were cleaned using a multi-stage wash process involving 30% acetone, 1% Triton X-100 solution, and Milli-Q water. Cleaned toenail samples were digested using an open vessel microwave assisted digestion method adapted from the Dartmouth Trace Element Analysis Core (Andrew et al. 2020). Briefly, samples were digested with 0.5 ml nitric acid (HNO_3, OptimaTM grade) and 0.2 ml of hydrogen peroxide (H_2O_2, OptimaTM grade) and heated to 110 °C before being diluted with 0.5% hydrochloric acid (HCl) for analysis by ICP-MS (8800 ICP-MS Triple Quad; Agilent technologies,

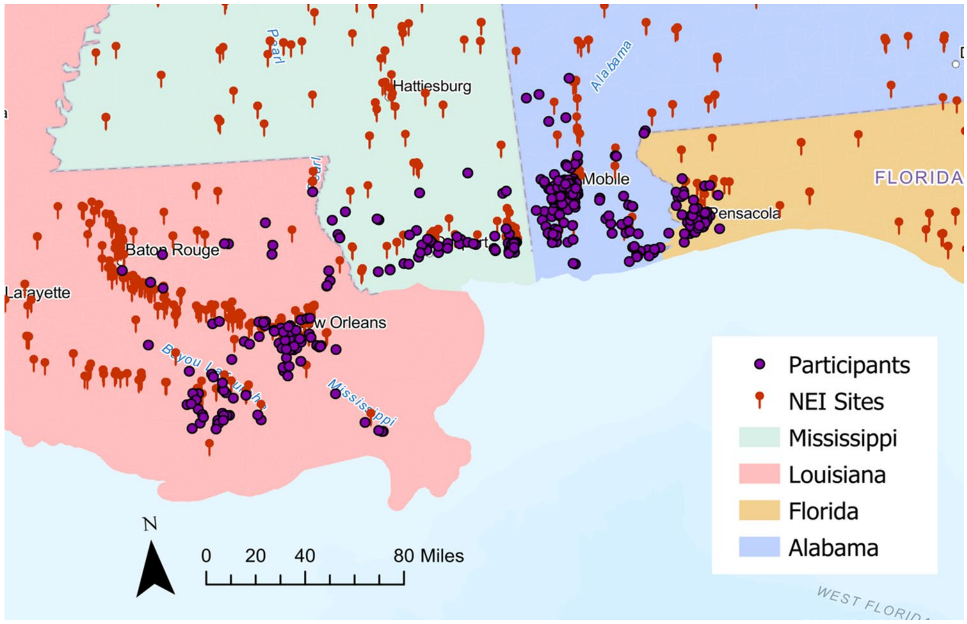

Fig. 1 Geographic distribution of participant residences and metal emitting NEI sites

Inc., Santa Clara, CA). Data quality was monitored via multipoint calibration curves for each analyte at the beginning and end of each batch, analysis of laboratory and digestion blanks, duplicates, spikes, and comparison with two reference materials: human hair Japan NIES #13 (National Institute for Environmental Studies, Ibaraki, Japan) and caprine horn NYS RM 1801 (New York State Department of Health Wadsworth Center, Albany, NY).

This analysis focused on elements that were both detected in > 85% of toenail samples and reported by the National Emissions Inventory (NEI) (As, Cr, Hg, Mn, Pb, Se). Nickel (Ni) was excluded despite high detection (100%) and NEI reporting because our previous reliability study found no correlation between toenail Ni concentrations from the same person over two time points ~ 3 years apart, suggesting that the toenail matrix may not be a good biomarker of long-term Ni exposure (Lin et al. 2023). The average between-batch coefficient of variation across metals was 11% and ranged from 3% (Pb) to 18% (Hg). The limit of detection (LOD) for each metal was calculated using 3 times the standard deviation of digestion blanks ($n = 7$) for each batch. The average LOD for each metal across batches ranged from 0.0003 µg/g for As to 0.0016 µg/g for Cr (Supplemental Table 1). Samples below the LOD (As, $n = 54$; Cr, $n = 29$; Pb, $n = 25$; Mn, $n = 8$; Hg, $n = 25$, Se, $n = 4$) were assigned a value of the batch-specific LOD divided by $\sqrt{2}$ (Helsel 2005).

National Emissions inventory

Sources of anthropogenic metal emissions were identified using the National Emissions Inventory (NEI), the US Environmental Protection Agency's (EPA) most comprehensive database of annual criteria, precursor, and hazardous air pollutant emissions (National Emissions Inventory 2014). Estimates provided by the NEI are compiled using reporting data provided by State, Local, and Tribal air agencies that are supplemented by information from the Toxics Release Inventory, the Acid Rain Program, and EPA's regulatory air toxics data. We abstracted all records of reported emissions of available metals (As, Cr, Hg, Mn, Pb, Se) from the 2014 NEI point source database and geocoded participant residential addresses relative to the locations of NEI sources to assess potential associations between residential NEI proximity and toenail metal concentration.

We assigned exposure to point sources of metal emissions using 3 different proximity metrics. First, we assessed linear distance measured as the Euclidean distance between the address of residence at the clinical exam and the nearest metal-emitting NEI site (Distance km). Second, we assessed an inverse distance weighted sum of the number of sites within a 30km radial buffer of the participants' residential address (Site IDW) using Eq. 1.

$$\sum_j \frac{I(d_{ij} < 30km)}{d_{ij}} \quad (1)$$

where d = distance indexed by i for the participant and j for the NEI site.

Third, we calculated the sum of the inverse distance weighted pounds of emissions within 30 km of the participants' residences (Emissions IDW) following the same logic as the Site IDW score. For each residential address, we summed the reported volume of emissions at each NEI site divided by the distance from the residence for all the NEI sites within a 30 km radius of the residence.

Statistical Methods

We used multivariable linear regression to estimate the difference in the $\log_{10}$-transformed toenail metal concentrations and 95% confidence interval (CI) per unit increase in each of the NEI proximity metrics (Distance km, Site IDW, and Emissions IDW scores). Models were adjusted for individual-level physiological or behavioral factors that could influence toenail metal concentration including age (years), cigarette smoking history (former/never), body mass index (BMI) (continuous), passive smoke exposure (> 30 min of smoke exposure per day on average), employment status (working, unemployed/retired), and state of residence. All individual-level covariate data were ascertained from the GuLF Study clinical exam and follow-up questionnaires closest to the time of toenail collection.

Analyses were stratified by self-reported race and income to assess potential disparities related to the effects of historic segregation and persistent racism in this area. Given the small proportion of participants in other racial categories, analyses of racial disparities focused on comparisons between the White and Black participant groups. As only 11 of the 413 participants in this study identified as Hispanic, we did account for Hispanic ethnicity.

In secondary analyses, we additionally adjusted for social factors such as individual level of educational attainment and neighborhood-level variables (median household income, percent of households below the poverty level, and median year that structures were built) using data from the 2014 American Community Survey (ACS) at the census block group level to address potential influence from unmeasured confounders related to residential proximity to industry. Social variables were each included in separate models to reduce the impact of collinearity. Beta estimates for all models were converted to percent differences using the formula $\left(10^{\beta} - 1\right) * 100$

Sensitivity Analyses

Toenail samples analyzed in this study were collected 2 – 6 years (median: 4.6) after the end of self-reported oil spill cleanup work and thus are well beyond the expected exposure window relevant to oil spill cleanup exposure. However, given the occupational origins of this cohort, we conducted sensitivity analyses including cleanup-related cumulative total average hydrocarbon (Cumulative THC; ppm) inhalation exposure estimates as a proxy of oil spill cleanup involvement and intensity in our models.

Results

Toenail metal concentrations of the metals of interest were not associated with oil spill cleanup involvement or timing of toenail sample collection relative to cleanup activity. Thus, metals reported in the toenail are interpreted to reflect non-oil spill related exposures.

Participant Characteristics

Forty-six percent ($n = 191$) of participants in this study self-identified as Black, 46% ($n = 190$) identified as White, and the remaining 8% ($n = 32$) identified as one of the following: Asian, American Indian/Alaskan Native, Mixed Race, or Other. Analyses for the 32 participants who identified as something other than White or Black were also excluded since there were very few participants in each of the racial sub-categories in this group. On average, White participants were older, had higher educational attainment, higher annual household income, were more frequently former smokers and currently employed compared to Black participants. There were also differences in racial makeup by state, with the majority (53%) of Black participants residing in Alabama and White participants having more even distribution across the 4 states (Table 1). Forty-nine percent of Black participants lived within 3 km of a metal-emitting NEI site compared to just 17% of White participants. Racial differences in residential proximity to metal-emitting NEI sites persisted after accounting for income, with Black participants living closer to metal-emitting NEI sites than White participants within every income category (Supplemental Fig. 1).

Industrial Determinants

We observed significant associations between NEI sources of metal exposure and toenail metal concentrations for Pb and Hg. No significant associations with NEI sites were observed for any of the remaining metals tested (As, Cr, Mn, and Se). Other personal and behavioral factors associated with toenail Pb and Hg concentrations are provided in Supplemental Table 2. Notably, toenail Pb concentrations were significantly inversely associated with participant education and neighborhood median structure age. Oil spill cleanup involvement was not related to toenail metal concentrations of any of the elements of interest in this study.

Lead (Pb)

Distance to the nearest Pb-emitting NEI site and density of NEI sites within 30 km of the residence (measured by

Table 1 Participant characteristics by self-reported race

	Total ($n=413$) n (%)	White ($n=190$) n (%)	Black ($n=191$) n (%)
Age			
20–39	120 (29)	42 (22)	71 (37)
40–59	222 (54)	100 (52)	104 (54)
60–69	71 (17)	48 (25)	16 (9)
Highest educational attainment			
< High school	80 (19)	30 (16)	36 (19)
High school or equivalent	149 (36)	61 (32)	83 (43)
Some college	124 (30)	53 (27)	61 (32)
> College graduate	60 (15)	46 (24)	11 (6)
Annual Household Income			
< $20,000	130 (31)	33 (17)	91 (48)
$20,000–$49,999	140 (34)	56 (29)	69 (35)
> $50,000	120 (29)	89 (46)	21 (11)
N/A	23 (6)	12 (6)	10 (5)
Smoking history			
Never	274 (66)	102 (54)	149 (78)
Former	139 (33)	88 (46)	42 (22)
Passive smoke exposure			
< 30 min/day	323 (78)	146 (77)	152 (80)
> 30 min/day	86 (21)	44 (23)	36 (19)
N/A	4 (1)	0 (0)	3 (1)
Employment status			
Working	253 (61)	136 (72)	96 (50)
Unemployed	123 (30)	31 (16)	85 (45)
Student/retired/other	37 (9)	23 (12)	10 (5)
State of residence			
AL	176 (43)	71 (38)	101 (53)
FL	55 (13)	33 (17)	15 (8)
LA	111 (27)	61 (32)	35 (18)
MS	70 (17)	24 (13)	39 (21)
Cumulative total average hydrocarbon (THC; ppm)			
Median (GSD)	91.6 (6.0)	85.4 (6.7)	102.2 (5.0)

the Site IDW score) were positively associated with toenail Pb concentration. For every 1 km increase in the distance from the home to the nearest Pb NEI site, we observed − 5.10% (95% CI: − 9.07, − 0.95) change in toenail Pb concentration after adjusting for personal and behavioral characteristics such as age, BMI, smoking history, passive smoke exposure, and employment status. Similarly, for every 1 unit increase in Site IDW score (higher NEI density around the home), we observed 64.5% (95% CI: 16.6, 132) higher toenail Pb concentrations after adjusting for personal and behavioral characteristics. Emissions volume from nearby NEI sites (measured by the Emissions IDW score) was positively related to toenail Pb concentration but the association was not statistically significant (Fig. 2).

Mercury (Hg)

Unexpectedly, proximity to Hg emitting NEI sites was inversely associated with toenail Hg concentration. For every 1 unit increase in Site IDW score (higher density), toenail Hg concentrations changed by − 47.4% (95% CI: − 66.9, − 16.4). We observed a similar trend with emissions volume. For every unit increase in Emissions IDW score (more emissions), we observed − 2.62% (95% CI: − 4.61, − 0.66) changes in toenail Hg concentration (Fig. 2). Distance to the nearest Hg NEI site was not significantly associated with toenail Hg concentration.

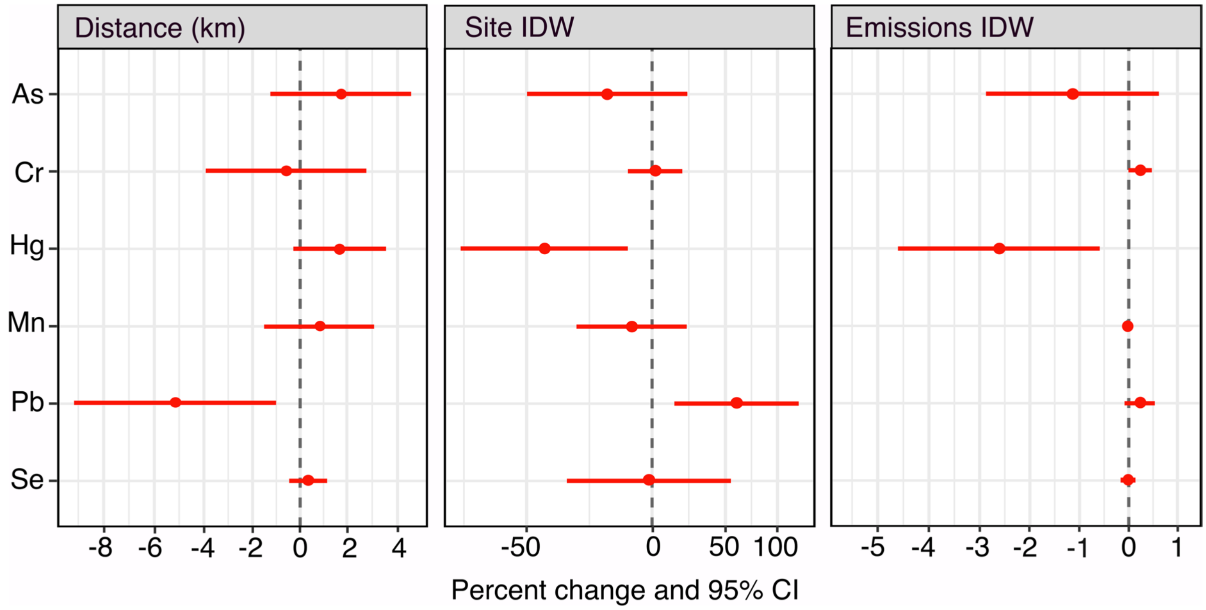

Fig. 2 Relationship between industrial determinants of metal exposure (Distance, Site IDW, Emissions IDW) and toenail metal concentrations adjusted for age, cigarette smoking history, body mass index (continuous BMI), passive smoke exposure (>30 mins of smoke exposure per day on average), employment status (working, unemployed/retired), and state of residence. Distance (km) = linear residential distance from the closest NEI site. Site IDW = inverse distance weighted number of sites within 30 km of residence. Emissions IDW = inverse distance weighted volume of emissions within 30 km of residence

Stratification by Race

Given the racial differences in residential proximity to NEI sites in this study, we stratified analyses by race to examine the potential inequities in metal exposure burden from metal-emitting NEI sites. The change in toenail Pb concentration associated with a one unit increase in Pb Site IDW score was stronger among Black participants (110%, 95% CI: 17.5, 275) compared to White participants (52.5%, 95% CI: – 11.6, 163). The relationships for Hg Site and Emissions IDW scores were similarly driven by stronger associations among Black participants. For every 1 unit increase in Hg Site IDW score, we observed – 72.1% (95% CI: – 88.5, – 32.5) change in toenail Hg among Black participants and – 3.52% (95% CI: – 47.1, 76.0) among White participants. Similarly, for every 1 unit increase in Hg Emissions IDW score, we observed – 4.70% (95% CI: – 8.17, – 1.09) change in toenail Hg among Black participants compared to – 0.81% (95% CI: – 3.29, 1.73) among White participants (Supplemental Fig. 2).

Secondary Analysis

Since industrial sites tend to concentrate in disadvantaged neighborhoods that may also experience disproportionate exposures to metals through other non-industrial sources, we additionally adjusted for individual-level and neighborhood-level socioeconomic variables to understand the extent to which observed associations could be reasonably attributed to metal exposures from unmeasured, non-NEI sources.

After additionally adjusting for individual-level educational attainment, distance from the nearest Pb-emitting NEI site and density of Pb NEI sites within 30 km of the residence remained significantly associated with toenail Pb concentration at -5.10% (95% CI: – 9.07, – 0.95) and 64.5% (95% CI: 16.6, 132), respectively. After adjusting for neighborhood-level social factors including the percent of residents living below the poverty line and the median year that structures are built within the census block group, the association remained in the expected direction but the effect was significantly attenuated (Fig. 3). Greater attenuation was observed after adjusting for the census block group median structure age (– 2.07%, 95% CI: – 6.83, 2.92) than after adjusting for census block group percent below poverty level (– 3.83%, 95% CI: – 8.00, 0.53). The same trends were observed for Pb Site IDW (Fig. 3).

Inverse associations between toenail Hg and Site/Emissions IDW scores remained after adjustment for individual level of education but were also attenuated after adjustment for neighborhood-level SES factors such as census block poverty rate and median year that structures were built (Fig. 3). Unlike for Pb, the attenuated inverse associations between Site IDW score and toenail Hg concentration remained statistically significant after adjustment for census block group poverty rate (– 39.8%, 95% CI: – 62.5, – 3.32). The Emissions IDW score variable for Hg also remained

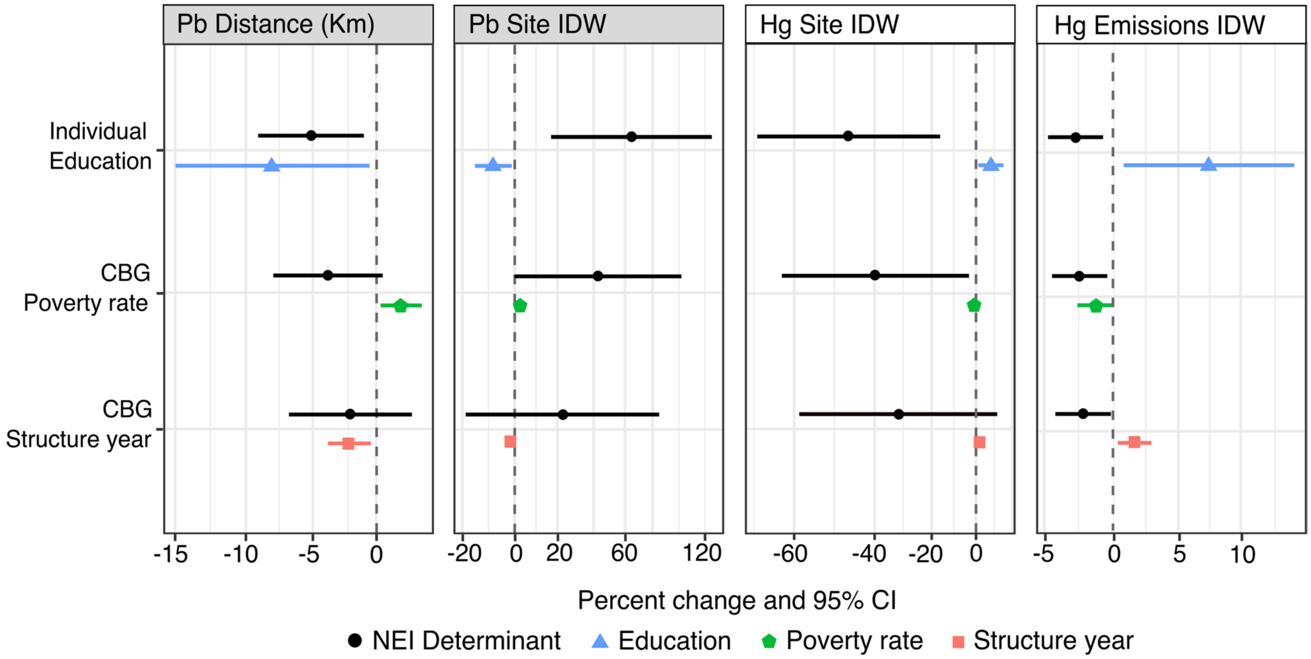

Fig. 3 Associations between significant toenail lead and mercury industrial determinants and toenail concentrations additionally adjusted for individual and neighborhood level SES variables (each in separate model). Individual Education = years of education from GuLF Study survey. CBG = census block group, data from 2014 ACS

statistically significant after adjusting for census block group poverty rate (– 2.45%, 95% CI: – 4.45, – 0.41) and the median year that structures were built within the census block group (– 2.20%, 95% CI: – 4.21, – 0.14).

Differences in Toenail Metal Concentration by Race

Despite greater proximity of Black residences near metal-emitting NEI sites, concentrations of all toenail metals tested except for Pb were significantly lower among Black participants (As (– 70.4%, 95% CI: – 83.1, – 48.3), Cr (– 46.8%, 95% CI: – 65.7, – 17.4), Hg (– 54.8%, 95% CI: – 69.7, – 32.6), Mn (– 47.8%, 95% CI: – 65.3, – 21.5), and Se (– 22.8, 95% CI: – 36.9, – 5.57) compared to White participants after adjusting for personal and environmental factors such as age, BMI, smoking history, passive smoke exposure, work status and state of residence. Median toenail Pb concentration was also lower among Black participants (– 28.3%, 95% CI: – 56.9, 19.3), but the difference was not statistically significant (Fig. 4).

We also examined differences in toenail metal concentration by income group and found that participants making less than or equal to $20,000 a year had toenail Pb concentrations that were 121% (95% CI: 18.7, 312) higher than those reporting making more than $50,000 a year. Significant differences in toenail metal concentration were also observed for Cr and Hg with those making less than $20,000 a year having toenail Cr and Hg concentrations that were

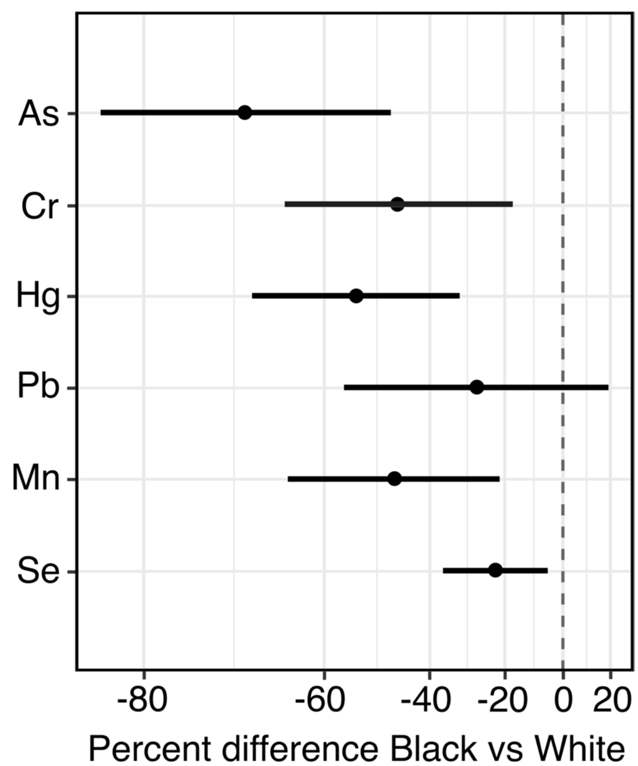

Fig. 4 Differences in toenail metal concentrations comparing Black participants to White participants adjusted for age, BMI, smoking history, passive smoke exposure, work status, and state of residence

45.7% (95% CI: 6.82, 68.4) and 64.7% (95% CI: 41.8, 78.5) lower, respectively, than those making more than $50,000 a year (Supplemental Fig. 3).

Comparisons with GuLF Study Blood Metal Measurements

To verify unexpected racial differences in metal body burden in this population, we examined blood metal concentrations from an existing GuLF study that collected whole blood samples from 1,058 participants at the home visit, 2 – 4 years prior to toenail sample collection. Blood samples were primarily collected for the assessment of oil spill cleanup-related benzene, toluene, ethylbenzene, and xylenes (BTEX) exposures but were additionally analyzed for Hg, Mn, Pb, and Se using ICP-MS. Blood concentrations of As and Cr were not measured. Among the 723 participants who were male and within the same age range as this study, we observed racial differences consistent with those observed using the toenail metal biomarker. Blood concentrations of Pb were not significantly different by race but concentrations of Hg, Mn, and Se were significantly lower among Black participants than White participants adjusting for age, BMI, smoking history, passive smoke exposure, and employment status (Supplemental Fig. 4).

Discussion

In this multi-state study of industrial metal exposures in the US Gulf, we found significant associations between residential proximity to NEI sites and toenail Hg/Pb concentrations as well as differences in residential proximity to NEI sites by self-reported race. Closer proximity and higher density of Pb-emitting NEI sites around the home were positively associated with toenail Pb concentration. On the other hand, residence farther from Hg NEI sites and exposure to lower volumes of NEI Hg emissions were associated with higher toenail Hg concentration. Despite greater residential proximity to metal-emitting NEI sites across every income category among Black participants, we found lower toenail concentrations of both toxic (As, Hg) and essential metals (Mn, Se, and Zn) in Black participants compared to White participants in this study.

Positive associations between residential proximity to Pb NEI sites and toenail Pb concentration were appreciably attenuated after the adjustment of census block group SES factors (percent of population below the poverty line and the median year that housing structures were built). There was also no association between inverse distance weighted volume of emissions and toenail Pb concentration suggesting that the relationship between residential proximity to Pb-emitting NEI sites may be, in part, driven by the fact that neighborhoods closer to metal-emitting sites are more likely to experience co-occurring exposures or other social stressors that may exacerbate their exposures to Pb. This phenomenon has been documented in previous studies in the US showing that industry-adjacent neighborhoods typically receive fewer public works maintenance or remediation projects, have older housing stock, and have limited bargaining power to prevent toxic environmental exposures from ending up in their communities (Geron et al. 2022; Tyrrell et al. 2013). In race-stratified analyses, we found stronger associations between proximity to metal-emitting NEI sites and toenail Pb and Hg concentrations among Black participants compared to White participants. Thus, consistent with known outcomes of historic redlining and other practices that promoted segregation, Black participants in this region may experience disproportionate burden of metal exposures from NEI sites. These findings add to a growing literature evidencing disproportionate distributions of industrial pollution burden among individuals from minoritized racial and ethnic groups in the US (Tyrrell et al. 2013; Mohai et al. 2009).

We suspect that dietary or non-spill cleanup-related occupational exposures, which were not well captured in the GuLF Study surveys, may explain the unexpected direction of metal exposure disparities observed in this study. Since the predominant sources of exposure to Hg, Mn, and Se are through the diet (Martins et al. 2020; Rose et al. 2010), differences in toenail concentrations of these elements may be attributable to dietary or nutritional differences across racial groups. As Hg in the toenail is largely comprised of methyl mercury (a common indicator of seafood intake) in non-occupational settings (Rose et al. 2010; Castro-González and Méndez-Armenta 2008), it is possible that environmental Hg exposure from ambient industrial exposures is masked by seafood intake related Hg exposures in this coastal population. As such, the relationship between Hg NEI proximity metrics and toenail Hg concentration in this study may reflect income-related seafood intake differences in this group with those living in higher SES neighborhoods, farther away from NEI sites, also consuming more seafood. The greatest sources of As exposure in the general population is through contaminated drinking water (Chung et al. 2014). Thus, it is possible that racial differences in toenail As concentration may be explained by differences in drinking water sources by neighborhoods of residence. On the other hand, Cr exposure is most often associated with occupational exposures or industrial processes (Sun et al. 2015; Wilbur et al. 2012). Studies with detailed occupational exposure data may be needed to explain racial differences in toenail Cr observed in this study.

This work expands the evidence supporting the use of toenail samples for metal exposure assessment. Specifically, we show the utility of toenail samples for capturing ambient Pb exposure trends. Significant inverse associations observed between toenail Pb concentration and the median year that structures were built within the residential census block group highlight a well-documented relationship between older housing stock and the greater exposure to Pb through outdated exposure sources such as lead paint or pipes (Hauptman et al. 2023). Racial differences in toenail concentrations of Hg, Mn, Pb, and Se were also corroborated by blood metal measurements in the GuLF Study. The consistency of metal exposure trends across matrices in the GuLF Study provides additional confidence for the reliability of the toenail metal biomarker, which has previously received pushback surrounding concerns about the lack of analytical standardization and potential for exogenous contamination (Gutiérrez-González et al. 2019). Among the metals unmeasured in GuLF blood samples, As has been validated as a biomarker of chronic exposure in the toenail (Slotnick and Nriagu 2006; Martinez-Morata et al. 2023). No studies have been conducted to validate toenail Cr as a biomarker of Cr exposure, but our previous toenail reliability study found strong agreement in Cr measurements across triplicate toenail samples thus providing analytical confidence in this measurement (Lin et al. 2023).

Compared to median concentrations reported in the National Health and Nutrition Examination Survey (NHANES) among White men of the same age range from the same period, median blood Hg concentrations from White GuLF Study participants were marginally higher, which may be reflect higher locally caught seafood consumption in Gulf states (Sathiakumar et al. 2017) (Supplemental Fig. 5). Blood concentrations of Mn, Pb, and Se from the GuLF Study were comparable to concentrations reported in NHANES (Supplemental Fig. 5).

A limitation of this work is the use of residential proximity from industry-reported air emissions sites and emissions volumes as the exposure metrics. There is no simple conversion of release quantity from NEI sites to the actual dose received by individuals since multiple processes can affect their fate and transport and determine how humans are eventually exposed to these pollutants (Maantay 2002; Brender et al. 2011; Huang and Batterman 2000). Furthermore, reporting to the NEI database is voluntary and designed for regulatory purposes. As such, data are limited to annual aggregate values and lack temporal or spatial variability. Another limitation is our use of socially constructed variables like race to delineate differences between groups, which may not perfectly capture differences in the way people experience environmental injustices. However, we do so in this study in efforts to describe the persistent effects of a long history of racial segregation and racist zoning laws in this region.

Limitations in emissions reporting and lack of detailed dietary/occupational information can muddy geospatial patterns of NEI metal exposures; however, our findings advance our understanding of environmental metal exposure assessment in two ways. First, consistency of metal exposure trends across biomarker matrices and positive associations between toenail Pb and neighborhood housing age provide substantial contributions to the validation of toenail samples for metal exposure assessment, specifically for Pb. Second, consistency in the of the directions of toenail Pb concentration and Pb NEI proximity across multiple proximity metrics and adjustments are suggestive of a positive contribution from Pb-emitting industries on Pb body burden in this region. These findings highlight the importance of prioritizing continued Pb mitigation interventions in industry-proximal neighborhoods where residents can be co-exposed to Pb from multiple sources that may have detrimental consequences on health and well-being.

Conclusion

This study identified racial disparities in residential proximity to metal-emitting NEI sites in the US Gulf region and highlighted unexpectedly higher toenail concentrations of both toxic (As, Hg) and essential (Cr, Mn, Se) metals in White participants compared to Black participants. The consistency in metal trends observed across exposure matrices and the confirmation of associations between toenail Pb with neighborhood housing age in the expected direction also provide additional confidence for the continued use of toenail samples for Pb exposure assessment on a population level. We highlight concerns about elevated Pb exposures in industry-proximate neighborhoods regardless of whether the exposures are coming directly from the facilities or from other factors related to industry-adjacent residence. Interventions to reduce metal exposure in this population should focus particular attention on disparities by race and income. Further studies focusing on diet and occupation should be conducted to pinpoint—and mitigate where appropriate—the sources of the unexpected metal exposure disparities in this population.

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Data Availability The data and code can be requested by email to the corresponding author.

Declarations

Conflict of interest The authors declare no competing interests.

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